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Article

The effect of pre-annealing on the evolution of the microstructure and mechanical behavior of aluminum processed by a novel SPD method

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Abstract: A novel continuous process of severe plastic deformation (SPD) called continuous close die forging (CCDF) is presented. The CCDF process combines all favorite advances of multidirectional forging and other SPD methods and it can be easily scaled up for industrial use. Keeping constant both the cross section and the length of the sample, the new method promotes a refinement of the microstructure. The grain refinement and mechanical properties of commercially pure aluminum (AA1050) were studied as a function of the number of CCDF repetitive passes and the previous conditioning heat treatment. In particular, two different pre-annealing treatments were applied. The first one consisted on a reheating to 350ºC for 1h aiming at eliminating the effect of the deformation applied during the bar extrusion. The second pre-annealing consisted on a reheating to 630ºC for 48h followed by a cooling at 66 °C / hour until reaching 300 °C, temperature at which the material remained for 3 hours prior to a final furnace cooling to room temperature, aiming at increasing the elongation and formability of the material. No visible cracking was detected in the workpiece of aluminum processed up to 16 passes at room temperature after the first conditioning heat treatment and 24 passes could be applied when the material was subjected to the second heat treatment. After processing through 16 passes for the low temperature pre-annealed samples, the microstructure was refined down to a mean grain size of 0.82 μm and the grain size was further reduced to 0.72 μm after 24 passes, applied after the high temperature heat treatment. Tensile tests showed the best mechanical properties after the high temperature pre-annealing and 24 passes of the novel CCDF method: yield strength and ultimate tensile strength of the ultrafine-grained aluminum were 180 and 226 MPa, respectively. Elongation to rupture was about 18%. The microstructure and grain boundary statistics are discussed with regard to the high mechanical properties of the UFG aluminum processed by this novel method.of about 200 words maximum.

Keywords: CCDF, UFG, EBSD, Mechanical properties, pre-annealing, Aluminum)

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1. Introduction

It is well known that the microstructure plays an important role in the physical and mechanical properties of polycrystalline materials. According to the Hall-Petch relationship, whichs describes the dependence between the yield stress, σγ, and the grain size, d, in equation (1) [1,2], the strength of metallic materials can be enhanced by grain refinement. During the last three decades, severe plastic deformation (SPD) methods have been successfully applied to achieve ultrafine-grained (UFG) materials, in the range between 100 nm and 1 μm, or even nanoscale structure in numerous pure metals and alloys [3,4]. Almost 20 years ago, the intention for a transfer of SPD to industry was disclosed [5,6]. However, no significant progress in that direction has occurred.

$$\sigma_y = \sigma_0 + k \cdot d^{-1/2} \quad (1)$$

If a SPD process is applied to a material, a UFG structure can be obtained with high angle grain boundaries. SPD processes introduce a great amount of strain in the material, and they must be performed at low temperatures in the presence of large hydrostatic pressures to avoid crack nucleation and propagation, which would compromise the integrity of the workpiece. Currently there are several SPD processes that meet the above specifications and generate uniform microstructures throughout the volume of the piece, which is important to have stability in the mechanical properties and subsequent forming processes.

The top-down SPD methods, have been proven to be an effective and promising alternative for the mass production of UFG materials and present advantages when compared to the bottom-up approach, based on nano-powder compaction. These include the lack of porosity, reduced levels of impurities and the possibility of scaling the process industrially, in terms of the process times and required investment [7,8]. In addition to the most widely explored SPD processes, HPT (High Pressure Torsion) and ECAP (Equal Channel Angular Pressing), MDF (Multidirectional forging) can also be scaled to industrial production [3-6]. The basic principle of MDF is the repetitive application of compression to a material, varying the axis of application of the deformation in each pass. The redundant plastic deformation will accumulate, after each pass, whether the deformation occurs at low or high temperature. The repetitive variation of the axis of application of the deformation is important for the refinement of the microstructure [3,8].

In this research work, a novel MDF method [9], named Continuous Closed Die Forging (CCDF), has been applied to a commercially pure aluminum (AA1050) with 99.5% purity. In addition to the new processing method, the material was subjected to two different pre-annealing treatments. The first pre-annealing treatment was performed at a low temperature for a short time and the second one at high temperature for a long time. According to the results, the new CCDF process was suitable to generate UFG microstructures regardless of the conditioning of the samples by different pre-annealing heat treatments. However, the pre-annealing conditions affected the number of deformation passes which could be applied to the samples and, thus, the final grain size and mechanical properties.

2. Materials and Methods

Bars of AW-Al-1050A with a diameter of 20 mm and a length of 100 mm were subjected to two different annealing heat treatments. The first annealing heat treatment was performed at 350ºC for 1h followed by air cooling. These conditions were selected in order to remove the effect of any strengthening generated during the extrusion process and homogenize the microstructure. The second pre-annealing consisted of a reheating to 630 °C for 48 hours, followed by a cooling at 66 °C / hour until reaching 300 ° C, temperature at which the material remained for 3 hours prior to a final furnace cooling to room temperature. This pre-annealing heat treatment has been reported to be effective in increasing the yield strength of 1050 aluminum alloy processed by ECAP [10,11]. The heat treatments were applied to the samples using an HOBERSAL 12 PR / 300 oven.

The principle of CCDF processing is shown in Figure 1. The tool (Figure 1a) is composed of two dies which form a rhomboidal inner cross section such as the one indicated in Figure 1b with a relationship between the major and minor axis of 3:1. The geometry has been defined in order to avoid tilting of the samples between deformation passes. The deformation sequence is described in Figure 1c, indicating that the samples, with an initial circular section of 20 mm in diameter, were placed in the cavity of the matrix and a closing load of 44 tones was applied to the dies through a rod, which was held for 10 seconds, forcing the sample to acquire the inner shape of the matrix. After the retraction of the rod, the dies were open and the sample piece turned 90 degrees counterclockwise for all the subsequent forming passes. In this new position of the sample, a closing load was again applied and the material flowed until it filled the space between the dies. The final shape of the samples after each deformation pass was rhomboidal with a major axis of less than 15 mm and a second axis which depends on the hardness of the material. Molybdenum disulfide lubricant was used in the process. The displacement speed of the rod was 5 mm/s and the temperature was controlled using a thermocouple and a data acquisition system. Following this CCDF route, up to 16 passes could be applied to the material which followed the low temperature pre-annealing condition and a total of 24 passes could be applied to the 1050 aluminum samples which followed the high temperature pre-annealing. It is worth mentioning that the length of the sample kept constant during all passes.

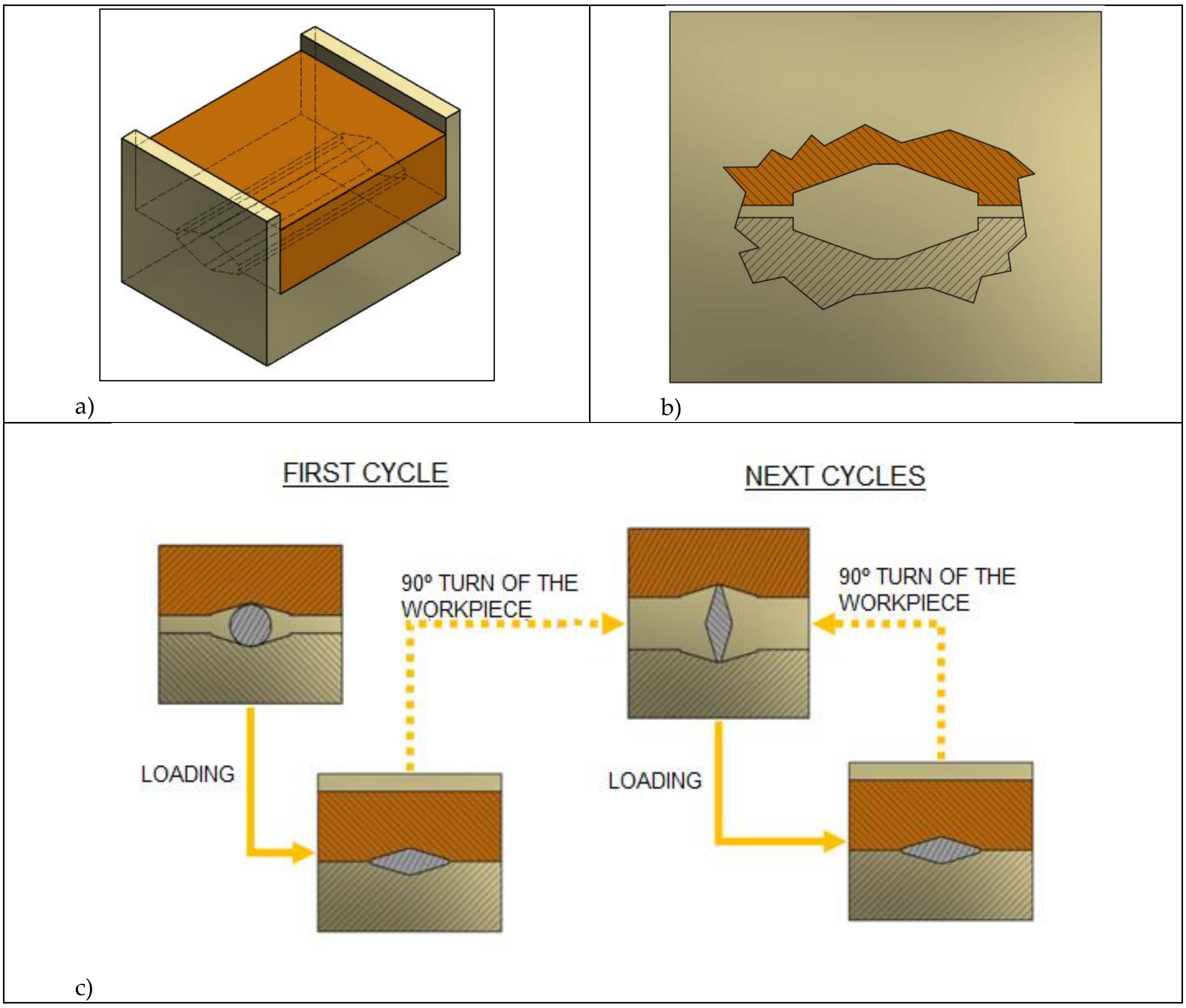


Figure 1. Schematic of continuous closed die forging (CCDF) including a) the matrix configuration, b) geometry of the dies and c) deformation sequence

For metallographic interpretation, sectioned samples were electropolished and the grain structures were recorded by orientation imaging microscopy (OIM) using the electron back scattered diffraction (EBSD) technique integrated in Scanning Electron Microscopy (SEM) brand JEOL, model JSM-5600 (controlled and analyzed using Channel 5 software). The grain size slightly differed between the samples, and different area dimensions and step sizes were chosen to maximize the number of data points and still get a good statistical results. The statistical variation on grain size and misorientation angle, obtained by EBSD, were used for the interpretation of the mechanical results. Additionally, the mechanical properties were determined on cylindrical microtensile samples machined from the bars after CCDF processing using a DEBEN microtensile testing machine, model MTEST5000S. The latter tests were carried out until the rupture of the sample.

3. Results and Discussion

3.1. Effect of the pre-annealing cycle on the microstructure after 8 passes

The microstructure generated in the cross sectional area, after the same number of passes, in this case 8 passes, was compared for a sample which had followed the pre-annealing at low temperature and a sample which had followed a pre-annealing at high temperature. Figure 2 represents the EBSD texture maps for both samples. In both cases, the material has undergone a refinement of the microstructure with the formation of small grains and subgrains, which exhibit a preferable orientation related to the orientation of the original grain generated during the pre-annealing treatment.

If EBSD data is analyzed in more detail, and the orientation of the grain boundaries is taken into account, small differences are detected for the different pre-annealing conditions. Figure 3 (a) and (b) represents the grain boundary maps for the low and high temperature pre-annealing, respectively. In this case, HABs (High Angle Boundaries) with misorientation angles larger than 15° are represented as black lines, while the LABs (Low Angle Boundaries) with misorientation angles lower than 15° are represented as green lines. In both cases, LABs are more common than HABs. The statistical analysis of the fraction of boundaries according to their misorientation is represented in Figure 3 (c) and (d) for the low and high temperature annealing, respectively. In this case, the fraction of LABs with the smallest misorientation represented in the graph is slighter higher for the sample pre-annealed at high temperature. However, when the boundaries are divided between those with a misorientation lower than 15° and the ones with a misorientation higher than 15° (Figure 3 (e) and (f), for the low temperature pre-annealing and high temperature pre-annealing, respectively), the results show that the high temperature annealing promotes more HABs, and thus new grains, than the low temperatures pre-annealing, which is related to a higher fraction of subgrains.

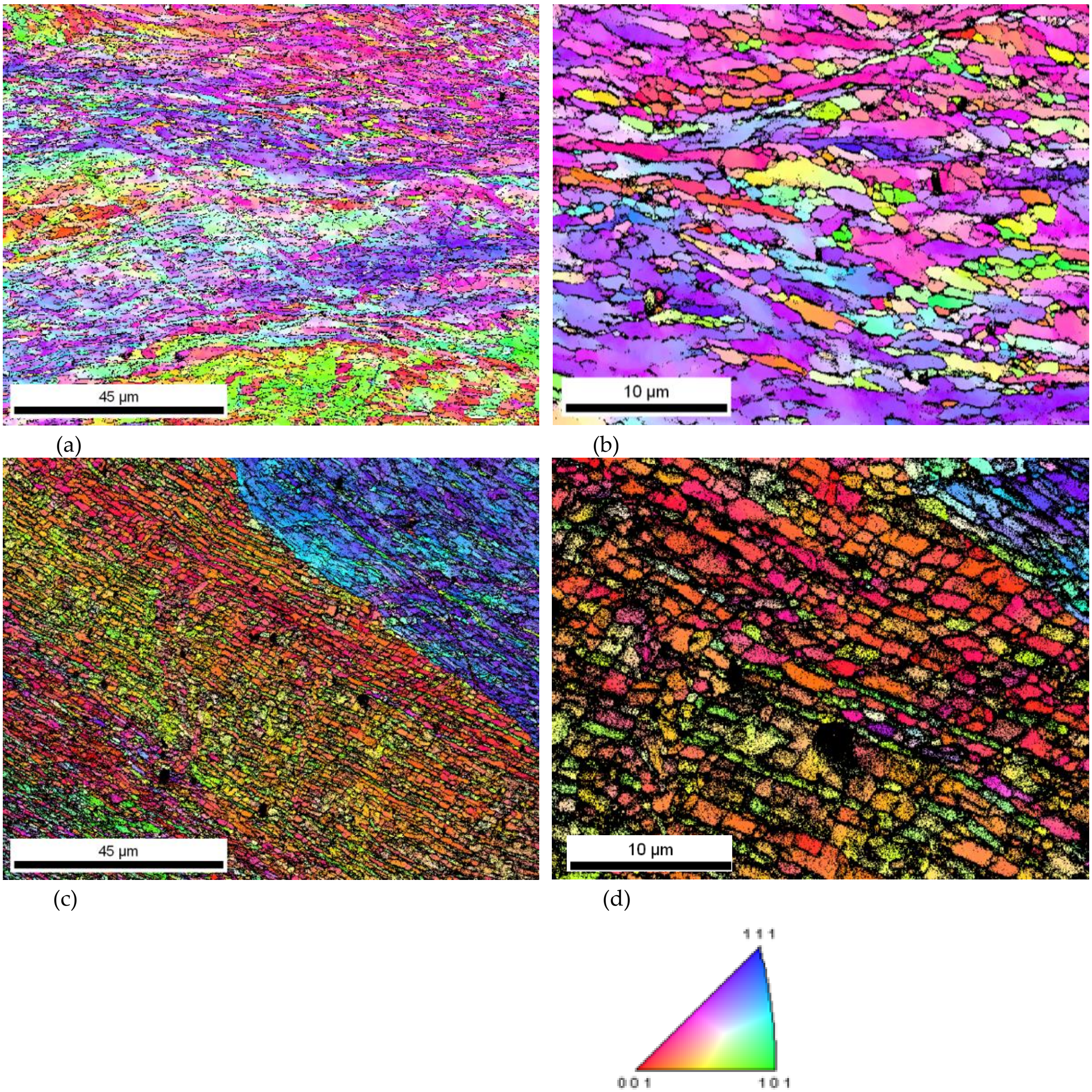
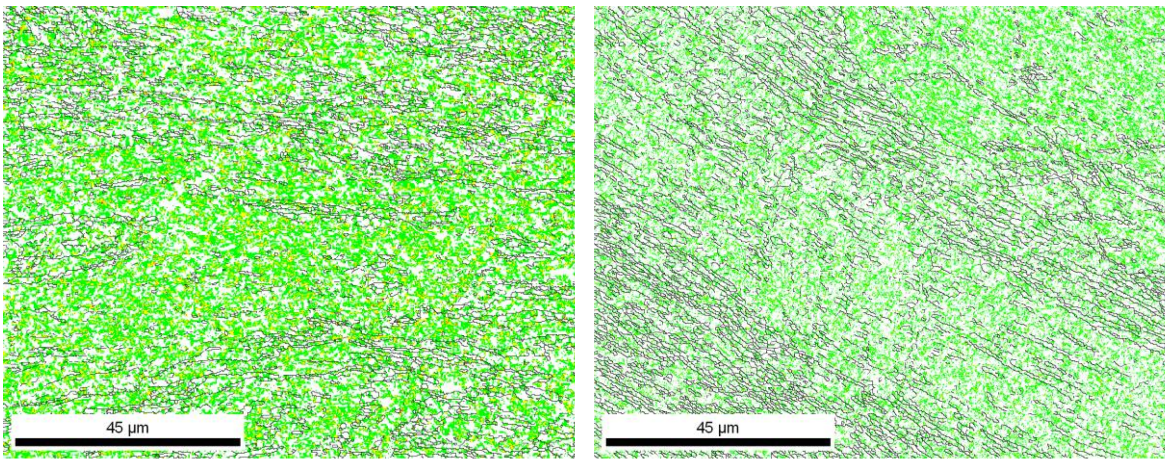


Figure 2. EBSD results after 8 passes of CCDF: Texture maps of (a) and (b) Al 1050 pre-annealing at low temperature and (c) and (d) Al 1050 after pre-annealing at high temperature



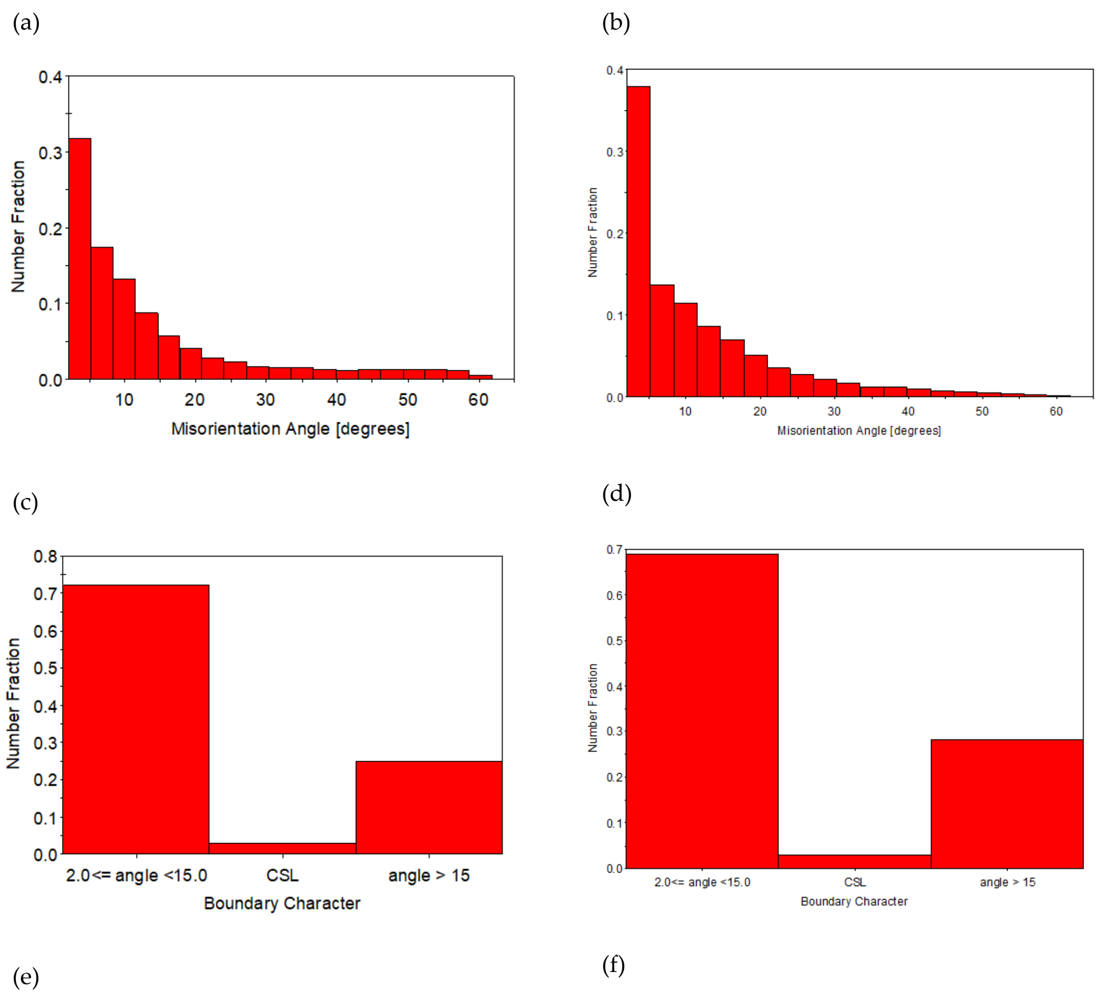
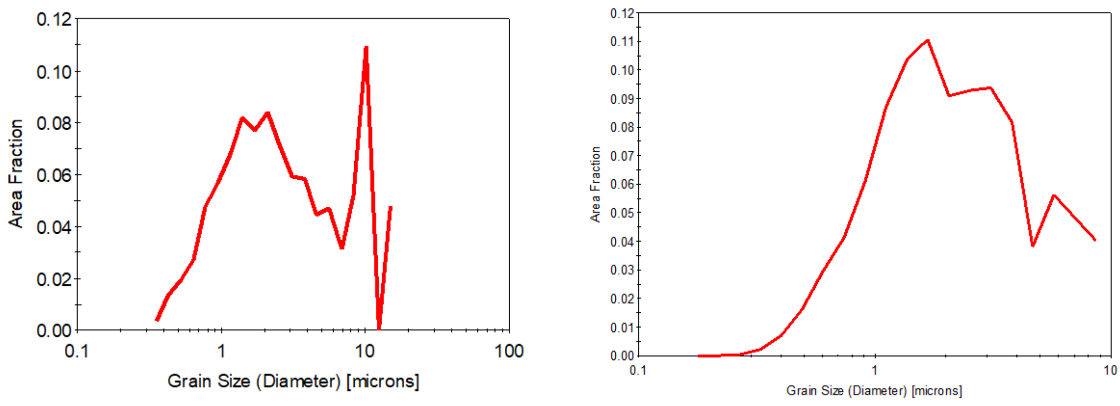


Figure 3. Grain boundary maps of Al 1050 with a pre-annealing at (a) low temperature and (b) high temperature, statistical analysis of boundaries misorientation for the samples with pre-annealing at (c) low temperature and (d) high temperature and quantitative analysis of LABs and HABs for a pre-annealing at (e) low temperature and (f) high temperature

Finally, the distribution of the grain size is represented in Figure 4 for both pre-annealing conditions. The distribution of the grain size is more homogeneous for the high temperature pre-annealing condition, although the distribution does not correspond to a UFG material for any of the cases, indicating that 8 passes is not enough, regardless of the pre-annealing treatment, to promote the desired refined microstructure for the current new CCDF process.

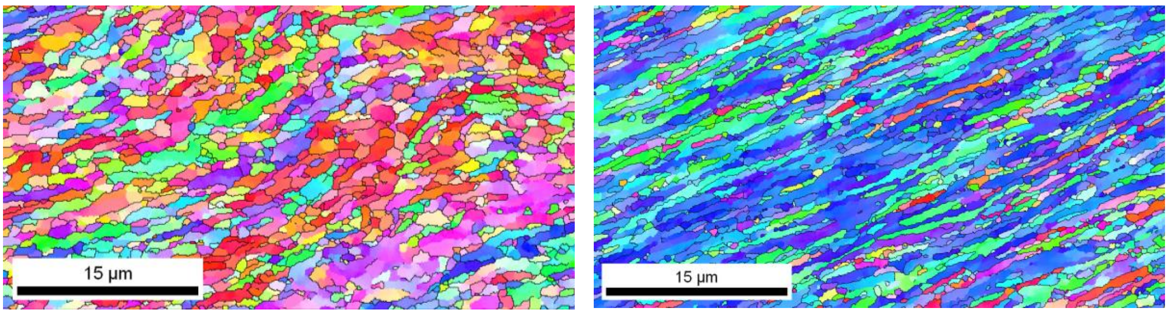


(a) (b)

Figure 4. Distribution of the grain size for (a) the sample pre-annealed at low temperature and (b) the sample pre-annealed at high temperature

3.2. Characterization of the microstructure at maximum number passes

The maximum number of passes which could be applied to the material, through the novel CCDF process, was 16 for the material pre-annealed at low temperature and 24 for the material pre-annealed at high temperature. Therefore, the first difference between the two pre-annealing treatments was the number of passes and accumulation of the deformation which could be applied, which were higher for the high temperature pre-annealing. Figure 5 represents the EBSD texture maps at the maximum number of passes for each pre-annealing condition. In both cases, the samples contain a mixture of elongated and equiaxed grains.



(a) (b)

Figure 5. EBSD texture maps for (a) sample subjected to a low temperature pre-annealing after16 CCDF passes and (b) sample subjected to a high temperature pre-annealing after 24 CCDF passes

If the characteristics of the grain boundaries is analyzed (Figure 6.) the increase in the fraction of HABs with respect to the microstructures after 8 passes is evident (Figure 3). However, there is still a high fraction of LABs, significantly higher after 16 deformation passes than after 24 deformation passes. Therefore, the possibility of increasing the amount of deformation which can be accumulated by increasing the pre-annealing temperature and time, promotes the formation of new grains and reduces the amount of subgrains. The misorientation distribution of boundaries obtained from the EBSD data and displayed in Figure 6 (c) and (d) correspond to a bimodal type with two different modes, low and high angles, as traditionally reported in the literature for highly deformed samples

[12-17]. Chang et al. reported that the dislocation density inside the grains of a 1050 aluminum alloy decreases with an increase of the strain, and most of the grains eventually became free of dislocations, due to a continuous dynamic recrystallization during the SPD processes after large deformations [18].

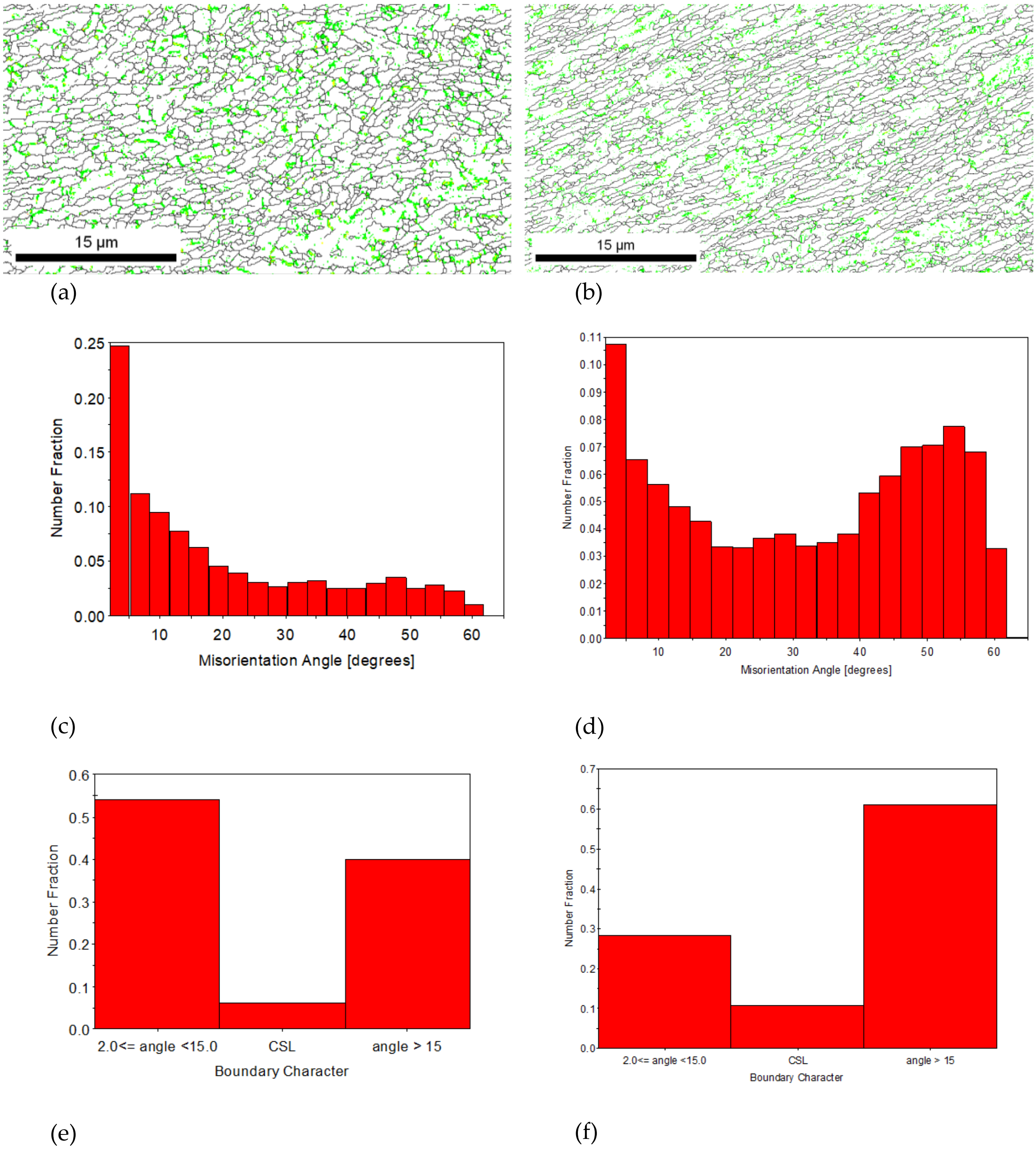
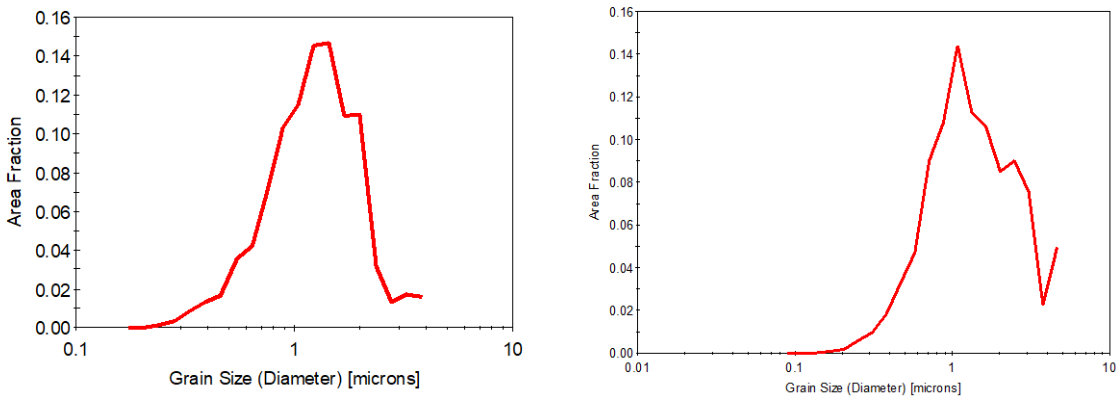


Figure 6. Grain boundary maps of Al 1050 after the maximum number of passes with a pre-annealing at (a) low temperature and (b) high temperature, statistical analysis of boundaries misorientation for the samples with pre-annealing at (c) low temperature and (d) high temperature and quantitative analysis of LABs and HABs for a pre-annealing at (e) low temperature and (f) high temperature

If the effect of pre-annealing on the grain size in taken into account, as represented in Figure 7, both pre-annealing conditions promote distributions which correspond to UFG materials. The EBSD data reveals that the average grain size has been reduced to 0.82 μm after 16 passes of CCDF for a sample pre-annealed at low temperature and to 0.72 μm after 24 passes of CCDF for a sample pre-annealed at high temperature for a long time. According to these results, it can be stated that the

CCDF process which has been proposed in this work as a novel SPD alternative, can effectively promote UFG microstructures.



(a) (b)

Figure 7. Distribution of the grain size after the maximum number of passes with a pre-annealing at (a) low temperature and (b) high temperature

3.3. Evolution of the mechanical properties

Figure 8 represents the Stress-Strain curves of the samples subjected to different pre-annealing treatment and deformation passes. When the curves corresponding to the pre-annealing at low and high temperatures are compared, it is evident how increasing the temperature and time of the pre-annealing promotes a curve with a higher UTS and the elongation is almost doubled. This increase in the elongation and ductility of the material, could explain the reason why the number of the deformation passes could be increased after the pre-annealing at high temperature.

After 16 CCDF passes, the microstructure exhibited the characteristics of UFG materials (Figure 5 and 7) and the stress-strain curves in Figure 8 show that the refinement of this microstructure can be related to an increase of the YS and UTS and a reduction of the elongation, regardless of the pre-annealing treatment. In fact, the uniform elongation (elongation up to the UTS) is severely reduced but the material exhibits high non-uniform elongation indicating that it is able to absorb energy during its plastic deformation. If the effect of the pre-annealing treatment in the stress-strain behavior of the material after 16 CCDF passes is analyzed, it can be inferred that the high temperature pre-annealing promotes higher UTS and elongation values and it is more effective when trying to optimize the mechanical properties promoted by the novel CCDF process. Therefore, pre-annealing at high temperatures not only promotes better mechanical properties for a constant number of deformation passes, but it also increases the formability, allowing to increase the total number of deformation passes which can be applied. This increase in the number of deformation passes promotes and extra increase of the UTS with small effect in the elongation to rupture.

In general terms, if pre-annealing at low temperature allows the application of 16 CCDF passes, related to a YS of 122MPa, a UTS of 156MPa and 12% of elongation to rupture, these properties can be increased to a YS of 180 MPa, UTS of 226 MPa and 18% elongation to rupture if the material is pre-annealed at high temperature and time and subjected to 24 deformation passes. The average increase of the mechanical properties is in excess of 40%. This confirms the results of previous works which indicate that an initial heat treatment prior to deformation at a relatively high temperature has a positive significant effect on the mechanical behavior of the material after SPD [11]. This condition is attributed to the ability of the material to withstand high deformations.

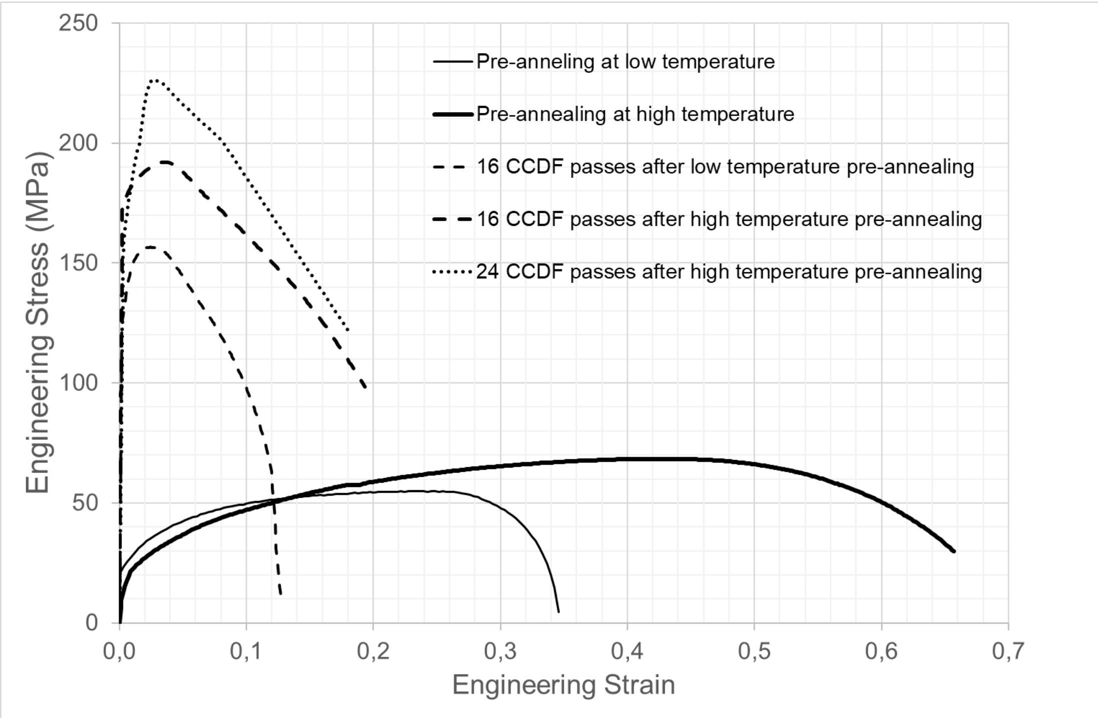


Figure 8. Stress–strain curves of Al-1050 after different pre-annealing and CCDF passes

4. Conclusions

Through a novel method of severe plastic deformation, called CCDF, an ultrafine grain size was obtained, which validates the concept. The material which has been used is a commercially pure 1050 aluminum alloy subjected to different pre-annealing conditions. The evolution of the microstructure is similar in the first deformation passes for which the material is not completely refined (a large substructure is still notice), but the processing window and mechanical properties are very sensitive to the initial conditioning of the material. When the material is pre-annealed a 350ºC for 1h, the maximum number of deformation passes is limited to 16, the average grain size is 0.82 μm and the mechanical properties are 122 MPa of YS, 156 MPa of UTS and 12% elongation to ruptures. These properties are in accordance with UFG materials but they can be further improved if the material is subjected to pre-annealing at 630ºC for 48h followed by a cooling to 300 °C, temperature at which the material remained for 3 hours prior to a final furnace cooling to room temperature. In this case, the total number of deformation passes could be increased to 24, promoting finer average grain size of 0.72 μm and the mechanical properties were increased by an extra 40%. This condition is attributed to the ability of the material to withstand high deformations.

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Conflicts of Interest: The authors declare no conflict of interest.

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